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COMPLETE SPECIFICATION

NO DRAWINGS

Polystyrene Foam having a High Content of Filler and Process for its Manufacture

We, KARL HOLL, a citizen of Germany, of Gorrestrasse 91, Heidelberg, Germany, JOSEF LINTNER, a citizen of Austria, of 25, Avenue des Bruyeres, La Garenne Colombes, Seine, France, HERBERT PETROVICKI, a citizen of Austria, of Heiligenbergstrasse No. 1-A, Heidelberg, Germany, FRIEDRICH SCHAFFERNAK, a citizen of Austria, of Monchbergweg 83, Heidelberg, Germany, and EDGAR ZEMB, a citizen of France, of 7, Rue des Marocains, Riedisheim, Haut-Rhin, France, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to foamed polystyrene articles and their production.

It is known that shaped articles can be manufactured from polystyrene which contains as blowing agent, for example, aliphatic hydrocarbons having boiling points between 0° to 100°C, such as light petroleum, by shaping it as with an extrusion press or by injection moulding, the resulting articles being expanded or foamed by heating. This method is, however, not suitable for the manufacture of all types of shaped articles in a smooth and technically simple manner, because in many cases some unknown property of the polystyrene containing the blowing agent causes changes in the article as first formed which result in deformation or distortion of the foamed article.

It has now been found that polystyrene containing a blowing agent can be converted in a technically advantageous manner and without the aforementioned disadvantages into articles of any desired shape, especially substantially two dimensional articles such for example as plates, ribbons or tapes, when the composition contains more than 10% (on the weight of the polystyrene) or fillers. No polystyrene foam having such a high content of fillers has been known in the past.

Accordingly the invention provides shaped articles of polystyrene foam containing at least 10%, preferably 10-100% (on the weight of the polystyrene) of a filler of average grain size of 1-50μ, preferably 1-20μ.

The shaped articles can be manufactured according to this invention by mixing granular polystyrene containing 2 to 6% of a vaporisable blowing agent, preferably one or more aliphatic or cycloaliphatic hydrocarbons having a boiling point of 0° to 100°C with filler extruding the mixture with an extrusion press or otherwise shaping it while maintaining the temperature at the exit of the press at a level such that the apparent density of the extruded article is less than half that of non-porous material of identical composition, cooling the extruded article to room temperature, and subsequently again heating it one or more times to complete the foaming or expansion.

The blowing agent can be incorporated in the polystyrene in any desired manner. For example granular polystyrene can be covered with a layer of petroleum ether, and the excess removed. The blowing agent can be added during the polymerisation of the styrene, or incorporated in the polystyrene with the aid of a liquid which is a solvent for both the polystyrene and the blowing agent.

The shaped articles are formed under pressure, preferably using an extrusion press or by injection moulding. When introducing the polystyrene containing the blowing agent into the press or the like, the required amount of filler can be added. This amount can vary within wide limits, depending on the end use of the foamed product. If it is

required that the product should be flexible and comparatively soft, a relatively small amount of filler will be used, say 10-20% of the weight of the polystyrene. If a harder, more rigid and tougher product is required, a larger amount of filler will be used. The nature and particle size and structure of the pigment all have an effect on the properties of the article.

It is immaterial whether the blowing agent is evenly distributed in all the grains of the polystyrene or whether individual grains have different contents of blowing agent. In the extreme case the polystyrene fed into the extrusion press may consist of polystyrene grains that contain blowing agent and others that do not, provided that the mixture as a whole contains 2 to 6% of blowing agent, on the weight of polystyrene.

The filler can be any finely divided inorganic or organic substance of average grain size about 1-50μ, preferably 1-20μ, which is insoluble in polystyrene and adequately thermo-stable.

Examples of suitable fillers are coloured and colourless water insoluble oxide and silicate pigments, and coloured organic pigments such as phthalocyanines and vat dyes of the anthraquinone series. Among inorganic pigments may be mentioned anhydrite, alumina, burnt magnesia, magnesite, silica gel and kieselguhr, titanium dioxide, and silicate pigments such as talc, fuller's earth, kaolin, bentonite, and asbestos flour. Coloured pigments include iron oxide, carbon black and chrome yellow. Mixtures of coloured and colourless pigments can be used.

According to their behaviour in the present process the fillers may be divided into two groups:

(a) Substances that give off a minor amount of gas or vapour under the extrusion conditions and act in polystyrene like chipped stones used as a boiling aid. When these products are used, some expansion takes place at the exit end of the extrusion press even at relatively low temperatures. These fillers are not incorporated in the plastic material as fillers in the conventional sense—the filled plastic material possesses the same physical properties, for example thermal stability, as a polystyrene foam which is free from fillers and has the identical polystyrene content. The filler has no influence on the mechanical properties of the material. These fillers produce expanded materials having the lowest specific gravity and very fine, regular pores. As examples of this group of fillers there may be mentioned kaolin, bentonite, iron oxide hydrates and the like.

(b) Fillers and pigments that are inert during the extrusion and are incorporated in the plastic material as fillers in the usual meaning of the word. To this group belong in particular calcined oxides, such as magnesium oxide, titanium oxide, antimony oxide and the like. To achieve adequate expansion at the exit end of the extrusion press there must be maintained within and at the exit end of the extrusion press—for a given mixture of synthetic resin and blowing agent and a given filler content—a higher temperature than is appropriate when a filler of group (a) is used. The pores produced are slightly larger than result from the use of the products mentioned under (a), and in this case the addition of the filler does have an influence on the mechanical properties of the plastic; for example its thermal stability is improved.

The filler used may, of course, also be a substance such as is used to render polystyrene flameproof; an example is the known combination of antimony oxide and an organic chlorine donor, for example a chloroparaffin. Particularly suitable organic chlorine donors are polyvinyl chloride, chlorinated polyvinyl chloride and polyvinylidene chloride, since—in contradistinction to the chloroparaffins—they do not impair the thermal stability of the polystyrene foam, but improve it. When antimony oxide is used in combination with an organic chlorine donor, it is advantageous to incorporate in the mixture also a filler of group (a).

Since many fillers, more especially those of inorganic origin, are opaque to infra-red radiation, a polystyrene foam filled with such substances not only has a lower specific gravity, but also a better thermal insulating power, than unfilled polystyrene foam of equal polystyrene content by weight.

When the filler is electrically conducting, such as a graphite powder or a conducting iron oxide or the like, the polystyrene foam becomes itself conductive. Such a material does not hold electrostatic charges, and also can be provided galvanically with a metal coating without a special surface pretreatment.

The compositions may be shaped and foamed by any conventional manner; e.g. an extrusion press or an injection moulding machine comprising a pre-plasticising device may be used.

The polystyrene foam obtained by the present process displays an unexpectedly even distribution of density and gives shaped articles, for example ribbons or tapes, having a surface that requires no after-treatment. Plates about 50 mm. thick and having at a kaolin content of for example 30% a dens-

ity less than 17 to 18 kg. per cu.m can be manufactured continuously. This corresponds to a polystyrene consumption of about 13 kg/cu.m The low density is achieved by subjecting the extruded article to several successive expanding operations.

If desired, the shaped article may be foamed or expanded in a heatable mould; in such a case the product takes the shape of the mould used. When a complicated shape is to be made, suitably cut pieces of material are placed in the mould; during the foaming process these melt together and then form a completely homogeneous object.

The following Examples illustrate the invention; all parts are by weight.

EXAMPLE 1

100 parts of granular polystyrene (average grain size 2 to 3 mm) containing about 4% of pentane are mixed with 20 parts of kaolin (average grain size about 5 microns) in a paddle mixer. This mixture is fed into an extrusion press comprising two screws (of diameter 77 mm, and length 7 diameters) and a ribbon die measuring 108×4 mm.

In the first zone of the press, where the material enters, the temperature is adjusted to 100°C, while the remainder of the machine is maintained at 125°C. The material is conveyed at a rate of 9 revolutions of the screw per minute. At the exit end of the die the ribbon has been expanded to a width of 200 mm. and a thickness of 12.5 mm. The density of the extruded material is about 168 kg. per cu.m. It leaves the press in the hot, plastic state and is conveyed over a mechanically driven stripper provided beyond the die and then for cooling, freely suspended, to the draw-off belt. It is then reeled on a drum of 3 metres diameter. It can be further expanded immediately, or after storage of for example 18 to 40 hours, in one or several stages. Any minor irregularities that may occur can be removed by means of a pressure roll. The ribbon can be cut up into pieces of the desired dimensions by means of a wire, which is advantageously heated electrically. After the final expansion the material weighs 22 kg. per cu.m.

In each of the following Examples the material after extrusion was treated as in Example 1 to complete the foaming or expansion.

EXAMPLE 2

100 parts of granular polystyrene (average grain size 2 to 3 mm) containing 7.5% of pentane, 100 parts of granular polystyrene free from blowing agent (average grain size about 0.5 mm) and 60 parts of kaolin (grain size about 5 microns) are mixed as described in Example 1 and the mixture is extruded as a ribbon.

Temperature in the feeding zone	117°C.
Temperature in the mixing zone	145°C.
Temperature in the ribbon die	125°C.
Density of the extruded ribbon	185 kg./cu.m
Density after final expansion	31 kg./cu.m
Thickness of polystyrene foam plates about	22 mm
Width of polystyrene foam plates about	410 mm

EXAMPLE 3

100 parts of polystyrene of average grain size 2-3 mm. and pentane content about 4%, and 50 parts of iron oxide of an average grain size of 10 microns, are mixed and extruded in ribbon form as described in Example 1.

Temperature in the feeding zone	117°C.
Temperature in the mixing zone	145°C
Temperature in the ribbon die	125°C.
Density of the extruded ribbon	186 kg./cu.m
Density after final expansion	38 kg./cu.m
Thickness of the polystyrene foam plates about	22 mm
Width of the polystyrene foam plates about	410 mm

The polystyrene foam is of a reddish brown colour.

EXAMPLE 4

100 parts of polystyrene of average grain size 2-3 mm. and pentane content 4%, and 30 parts of bentonite of a grain size of about 5 to 50 microns, are mixed and extruded in ribbon form as described in Example 1.

Temperature in the feeding zone	107°C.
Temperature in the mixing zone	130°C.
Temperature in the ribbon die	115°C.
Density of the extruded ribbon	102 kg./cu.m
Density after final expansion	26 kg./cu.m
Thickness of the polystyrene foam plates about	22 mm
Width of the polystyrene foam plate about	144 mm

EXAMPLE 5

100 parts of polystyrene of average grain size 2-3 mm. containing about 4% of pentane, and 30 parts of magnesium oxide of average grain size 5 microns, are mixed as described in Example 1 and extruded at the temperatures shown below:—

(a) temperature in the feeding zone	110°C.
temperature in the mixing zone	115°C
temperature in the ribbon die	125°C.
Density of the extruded ribbon	920 kg./cu.m
Density after final expansion	650 kg./cu.m
(b) temperature in the feeding zone	113°C.
temperature in the mixing zone	143°C.

temperature in the ribbon die 140°C.
Density of the extruded ribbon 323 kg./cu.m
Density after final expansion 46 kg./cu.m

The temperature used in run (a) was too low to give the results of the invention; the ribbon obtained is only slightly porous and cannot be formed into a light foam by the subsequent heating step. By contrast, at the temperatures used in run (b), the extruded ribbon is converted into a light foam by the subsequent heating operation.

EXAMPLE 6

100 parts of polystyrene of average grain size 2-3 mm. containing about 4% of pentane, and 30 parts of titanium dioxide of an average grain size of about 0.3 micron are mixed and extruded in ribbon form as described in Example 1.

Temperature in the feeding zone 110°C.
Temperature in the mixing zone 140°C.
Temperature in the ribbon die 130°C.
Density of the extruded ribbon 1103 kg./cu.m
Density after final expansion 1103 kg./cu.m

The extruded material could not be expanded, owing to the small particle size of the titanium dioxide filler.

EXAMPLE 7

100 parts of polystyrene of an average grain size of 2-3 mm. containing about 4% of pentane, 30 parts of kaolin (see preceding Examples), 10 parts of antimony oxide and 10 parts of after-chlorinated polyvinyl chloride containing about 70% of chlorine, are mixed and extruded in ribbon form as described in Example 1.

Temperature in the feeding zone 115°C.
Temperature in the mixing zone 142°C.
Temperature in the ribbon die 125°C.
Density of the extruded ribbon 158 kg./cu.m
Density after final expansion 22 kg.cu./m
Thickness of the polystyrene foam plate 28 mm
Width of the polystyrene foam plate 440 mm

The resulting polystyrene foam is only slightly flammable; if the material ignites, the flame is automatically extinguished shortly after removal of the igniting flame; in other words it is "self-extinguishing".

WHAT WE CLAIM IS:—

1. Shaped articles of polystyrene foam containing at least 10% (on the weight of the polystyrene) of a filler of average grain size of 1-50μ.

2. Shaped articles according to claim 1, wherein the grain size of the filler is 1-20μ.

3. Shaped articles according to claims 1 or 2, which contains 10-100% (on the weight of the polystyrene) of filler.

4. Shaped articles according to any one of the preceding claims, wherein a flame-proofing filler or mixture of fillers is used.

5. Shaped articles according to claim 4, wherein a mixture of antimony oxide with polyvinyl chloride, chlorinated polyvinyl chloride, or polyvinylidene chloride is used as filler.

6. Shaped articles according to any one of claims 1 to 4, wherein the filler is an electrically conducting filler.

7. Shaped articles according to claim 1, substantially as hereinbefore described.

8. Shaped articles according to claim 1 substantially as hereinbefore described with reference to any one of the Examples.

9. Process for the production of shaped articles claimed in any one of the preceding claims, which comprises mixing polystyrene containing 2-6% of a vaporisable blowing agent with the filler shaping the mixture in a press at a final temperature such that the extruded article has a density less than half that of the same material in the non-porous state, cooling the article to room temperature, and subsequently further expanding it by heating.

10. Process according to claim 9, wherein the blowing agent is an aliphatic or cyclo-aliphatic hydrocarbon or mixture of these hydrocarbons having a boiling point of 0 to 100°C.

11. Process according to claims 9 or 10, wherein a mixture of polystyrene granules having a high content of blowing agent, with polystyrene granules having a lower content or free from blowing agent is used.

12. A process according to any one of claims 9 to 11, wherein the shaped article is subjected to a plurality of successive expanding operations by alternately cooling and heating it.

13. A process according to any one of claims 9 to 12, wherein the shaped article is expanded in a mould.

14. A process for the manufacture of shaped articles of polystyrene foam according to claim 9 substantially as hereinbefore described.

15. A process for the manufacture of shaped articles of polystyrene foam according to claim 9 substantially as hereinbefore described with reference to any one of the Examples.

16. Shaped articles of polystyrene foam obtained by a process claimed in any one of claims 9-15.

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